

Variability of the Phase Function of Atmospheric Aerosols at Large Scattering Angles

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(Manuscript received 11 April 1996, in final form 10 March 1997)

ABSTRACT

The variability of phase functions of submicrometer atmospheric aerosols was studied using Mie theory. In the analyzed statistical ensemble of phase functions, the effective radii of particles were 0.1–1.5 μm , real parts of refractive indices were 1.45–1.6, imaginary parts of refractive indices were 0.001–0.01, and coefficients of variance of particle size distributions were 0.2–1.1. An ensemble average, a coefficient of variance of a phase function, and a skewness were calculated.

It was found that the coefficient of variance of a phase function has a minimum near a scattering angle of 150° . This fact was used to estimate the error in the aerosol optical thickness determination from space in the single scattering approximation of the radiative transfer theory.

1. Introduction

Atmospheric aerosols are of considerable importance in different fields of modern science and technology. Their physical and chemical properties have been studied by many authors (Junge 1963; Twomey 1977; Whitby 1978; d'Almeida et al. 1991). In particular, it was recognized that there are three distinctive modes of aerosol matter:

- nucleation mode (0.001 to 0.1 μm);
- accumulation mode (0.1 to 1 μm); and
- coarse particles mode ($>1 \mu\text{m}$).

The nucleation mode is produced by the gas-to-particle conversion; the accumulation mode is produced by a coagulation and a heterogeneous condensation; and the coarse mode has an origin in mechanical processes. The light-scattering efficiency of the nucleation mode is very low, so the phase function $p(\theta)$ of the atmospheric aerosol depends on the accumulation and coarse particle modes (Shifrin and Zolotov 1996).

The phase function at small scattering angles depends mostly on large particles ($a \gg \lambda$, where a is the radius of a particle and λ is the wavelength). The accumulation mode (along with the molecular scattering) is responsible for the phase function behavior at large scattering angles θ . This is determined by the fact that the number concentration of large particles N_l is very small in com-

parison with the number concentration of small particle N_s . According to Radiation Commission (1986), the ratio $\gamma = N_l/N_s$ is about 10^{-6} . In addition, it should be pointed out that phase functions of small particles are greater than or nearly equal to values of phase functions of large particles at scattering angles exceeding 90° . The phase function of the atmospheric aerosol at scattering angles exceeding 90° is of particular interest for many applications (e.g., climate forcing, satellite remote sensing, atmospheric correction problems).

The task of this paper is to investigate this function in detail using a statistical approach. The coefficient of variance, the skewness, and the mean for the statistical ensemble of phase functions were calculated at effective radii of $a_{ef} = 0.1\text{--}1.5 \mu\text{m}$, a coefficient of variance of the particle size distribution (PSD) of $\Delta = 0.2\text{--}1.1$, real part of the refractive index of $n = 1.45\text{--}1.6$, and imaginary part of the refractive index of $k = 0.001\text{--}0.01$. These calculations were made using Mie theory (Shifrin 1968; van de Hulst 1981).

The results obtained were applied to estimating the error of the aerosol optical thickness determination from reflection function measurements (Koepke and Quenzel 1979; Quenzel and Kaestner 1980; Rao et al. 1989; Stephens 1994) at a scattering angle of 150° , where the coefficient of the variance of aerosol phase function has a minimum. Note that this angle is typical for satellite observations (Mishchenko et al. 1996). To simplify the problem, the single scattering approximation of the radiative transfer theory was used (Hansen and Travis 1974; Liou 1992; Stephens 1994). This approximation works well for sufficiently small optical depths and non-absorbing aerosols, typically the case over the open ocean (Gordon 1997).

The investigations in this paper are closely related to

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the studies of the effect of the aerosol variation on the radiance in the earth's atmosphere–ocean system. This problem was studied by Plass and Kattawar (1972) and Koepke and Quenzel (1979). These papers are more complete because they account for multiple light scattering and the reflection of light from the ocean surface. However, in the first paper, only aerosol concentration changes with a height at a fixed PSD were considered. In the second paper, only four types of the PSD of the Sahara dust aerosol were taken into account.

In this paper, about 300 PSDs were used to study the variation of the reflected light with the variation of microphysical characteristics of atmospheric aerosols in the single scattering approximation.

2. Statistical analysis of phase functions of the submicrometer aerosol

a. Particle size distribution

The phase function of atmospheric aerosols depends on the particle size distribution $f(a)$, the complex refractive index of particles $m = n - ik$, the shape, and the internal structure of particles. The PSD of spherical polydispersions can be represented by a lognormal distribution (Whitby 1978; Radiation Commission 1986; d'Almeida et al. 1991):

$$f(a) = \frac{1}{a \ln \sigma_g \sqrt{2\pi}} \exp\left(-\frac{\ln^2 a/a_g}{2 \ln^2 \sigma_g}\right), \quad (1)$$

where a is the particle radius, and a_g and σ_g are the geometrical mean radius and the geometric standard deviation of the PSDs.

A very important fact is that different spherical polydispersions (even multimodal ones) with different PSDs but with similar effective variance $v_{ef} = \langle a^2(a - a_{ef})^2 \rangle / \langle a^2 \rangle a_{ef}^2$ [or the coefficient of variance (CV) $\Delta = \sqrt{v_{ef}}$] and effective radius $a_{ef} = \langle a^3 \rangle / \langle a^2 \rangle$ have similar optical properties (Hansen and Travis 1974). Here $\langle \dots \rangle$ means averaging on the PSD.

The effective radius is proportional to the ratio of the average volume to the average surface of particles and has a fundamental importance in the atmospheric optics. The effective variance is the ratio of the variance of the PSD to the squared mean radius. The coefficient of variation Δ (which equals standard deviation/mean) is generally better understood intuitively than the effective variance. So this value, along with the effective radius, will be used in this paper to characterize a PSD.

It should be pointed out that the value of the standard deviation s is usually less than the mean radius $\langle a \rangle$ and $\Delta \leq 1$ for aerosol PSDs (Whitby 1978). Note that, according to the Bienaume–Chebyshev inequality, the probability of a particle having a radius a beyond the range $\langle a \rangle(1 \pm 3\Delta)$ is low. It is less than 0.003 for normal PSDs.

The following range of the PSD parameters will be studied in this paper: $a_{ef} = 0.1\text{--}1.5 \mu\text{m}$, $\Delta = 0.2\text{--}1.1$.

It is very unusual for the accumulation mode particles to have values of coefficients of variance Δ and effective radii a_{ef} outside these limits. Both smaller and larger particles have very short lifetimes (Junge 1963; Stephens 1994).

In some cases, the accumulation mode aerosols are a mixture of particles with different optical constants and PSDs. Only the case of a unimodal aerosol with fixed optical constants will be considered here. The problem of a mixture of different PSDs and optical constants of particles is beyond the scope of this paper. This problem is still unresolved. However, in some cases, the spectral reflectance of a bimodal lognormal distribution can be simulated very well with spectral reflectance of a single lognormal distribution with an appropriate radius and coefficient of variance (Tanre et al. 1996). Thus, some of our conclusions may have a wider applicability.

b. Variation of the effective radius and the coefficient of variance of the PSD

The statistical approach was used to study the variability of a phase function of a submicrometer aerosol at large scattering angles. The following statistical characteristics were calculated using Mie theory:

$$\langle p(\theta) \rangle = \frac{1}{n} \sum_{i=1}^n p_i(\theta), \quad (2)$$

$$s_p^2(\theta) = \frac{1}{n-1} \sum_{i=1}^n (p_i(\theta) - \langle p(\theta) \rangle)^2, \quad (3)$$

$$\gamma_p(\theta) = \frac{1}{(n-1)s_p^3(\theta)} \sum_{i=1}^n (p_i(\theta) - \langle p(\theta) \rangle)^3 \quad (4)$$

at scattering angles greater than 90° . Here $\langle p(\theta) \rangle$ is the mean phase function of the accumulation mode, $s_p^2(\theta)$ is the variance, and $\gamma_p(\theta)$ is the skewness. The following normalization condition for the phase function was used:

$$\frac{1}{2} \int_0^\pi p(\theta) \sin \theta d\theta = 1. \quad (5)$$

The statistical ensemble of phase functions under investigation $p_i(\theta)$ [see Eqs. (2)–(4)] was formed at $\lambda = 0.7 \mu\text{m}$, $m = 1.53 - 0.006i$ (Radiation Commission 1986) and $\Delta = 0.7$. The value of the effective radius was changed from 0.1 to 1.5 μm in 0.005 μm steps. About 300 phase functions thus formed the background of the statistical analysis.

The results of this computation are presented in Figs. 1a and 1b. The coefficient of variance $\Delta_p(\theta) = s_p(\theta) / \langle p(\theta) \rangle$ changes from 17% at $\theta = 90^\circ$ to 36% at $\theta = 180^\circ$ with the minimum around a scattering angle $\theta = 147^\circ$, where the CV of the phase function is only about 8% (see Fig. 1a). The coefficient of variance of the phase function is less than 15% from $\theta = 135^\circ$ to 153° . It should be pointed out that the CV of the phase function grows more rapidly in the direction of the backscattering (see Fig. 1a).

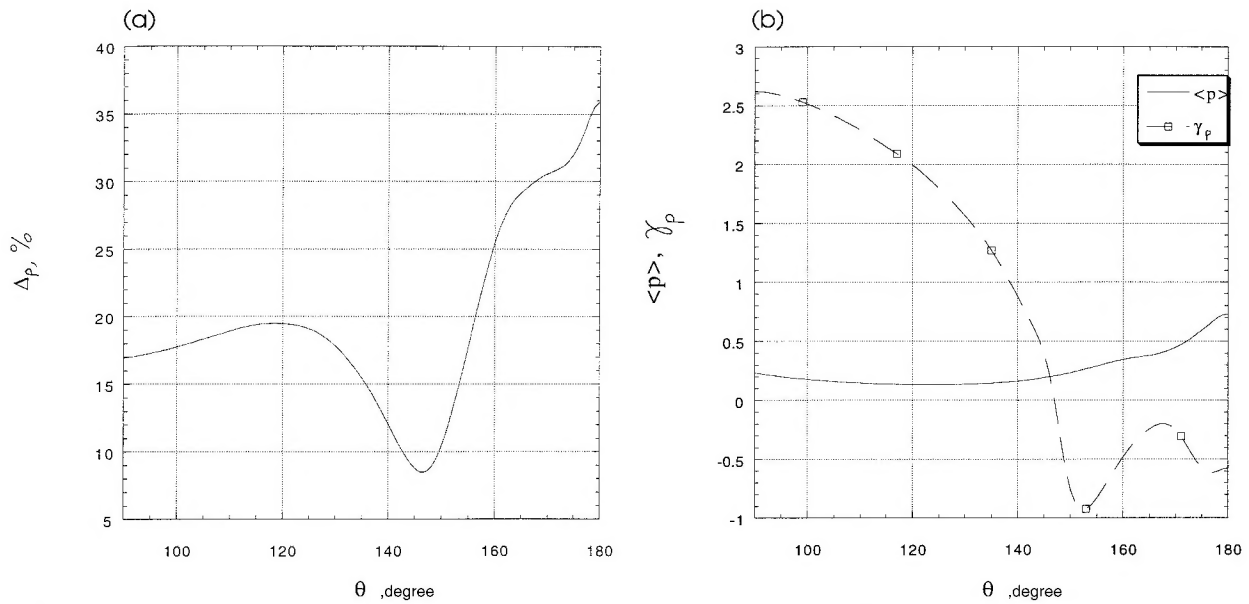


FIG. 1. Dependence of the coefficient of variance of phase function $\Delta_p = s_p(\theta)/\langle p(\theta) \rangle$ [see Eqs.(2) and (3)] on the scattering angle θ at $\lambda = 0.7 \mu\text{m}$, $m = 1.53 - 0.006i$, and $\Delta = 0.7$, (a) for different values of effective radii $a_{ef} = 0.1\text{--}1.5 \mu\text{m}$. (b) For the mean aerosol phase function $\langle p \rangle$ and the skewness γ_p .

It was found that there is a minimum of the coefficient of variation of the degree of polarization at a scattering angle of $\theta = 147^\circ$ as well.

In Fig. 1b, other characteristics of the statistical ensemble under consideration are represented. One can see that the skewness is positive for scattering angles of less than 147° . It is negative for $\theta > 147^\circ$. So the skewness changes a sign at $\theta = 147^\circ$, where the coefficient of variation of the aerosol phase function has a minimum (see Fig. 1a).

To study the dependence of the phase function on the coefficient of variance of the PSD, Δ , phase functions of aerosol media with different values Δ were calculated at $m = 1.53 - 0.006i$ and $a_{ef} = 0.1\text{--}1.5 \mu\text{m}$. Results for scattering angles of 90° , 120° , 150° , and 180° are presented on Figs. 2a–d, respectively. One can see that the phase function at $\theta \geq 90^\circ$ does not depend strongly on the value of the CV of the PSD [especially for values of $\Delta \geq 0.5$, which are quite usual for atmospheric aerosols (Whitby 1978)].

The results presented in Figs. 2a–d can be used to explain the minimum near $\theta = 150^\circ$ in Fig. 1a. The behavior of the phase function at $\theta = 150^\circ$ (see Fig. 2c) is like a sinusoid with an average value around 0.22 and a maximum deviation of the phase function from the average value of about 20%–30% (10%–15%, if the most representative values of $\Delta \geq 0.5$ is considered). At the same time, one can see that there is a monotonically decreasing (at the scattering angles 90° and 120°) or increasing (at the scattering angle 180°) phase function with the growth of particles. This is the reason for the minimum near the scattering angle of 150° in Fig. 1a.

Note that this feature is in accordance with the well-known fact (Hodkinson 1963) that phase functions of large particles at side scattering ($90^\circ\text{--}140^\circ$) are smaller than that of small aerosol particles, and vice versa at a scattering angle $\theta = 180^\circ$. The crossing point of different aerosol phase functions thus should be in the region of scattering angles from 140° to 160° . It is near 150° , as it can be seen from Fig. 1a. It should also be pointed out that phase functions around a scattering angle of 150° depend only slightly on shapes of aerosol particles (Hodkinson 1963; Mishchenko 1996).

c. Variation of optical constants

Let us now consider the variation of the phase function of atmospheric aerosol with the variation of the refractive index of particles.

It is well known that submicrometer aerosols are dominated by sulfates (Whitby 1978). In this case, the value of the complex refractive index is about $1.53 - 0.006i$ in the visible region of the spectrum (Radiation Commission 1986). It should be pointed out that the value of m depends on the humidity, the origin of the aerosol particles, their age, etc. Nevertheless, in most cases the real part of the refractive index of the submicrometer aerosol changes from 1.45 to 1.6, and the imaginary part, from 0.001 to 0.01 (Ivlev 1986).

To consider the variability of the phase function in this case, the statistical parameters, determined by Eqs. (2)–(4), were calculated for $a_{ef} = 0.2 \mu\text{m}$, $\Delta = 0.7$, and $\lambda = 0.7 \mu\text{m}$. The real part of the refractive index was changed from 1.45 to 1.6 in 0.001 steps with the imaginary part of the refractive index fixed ($k = 0.006$).

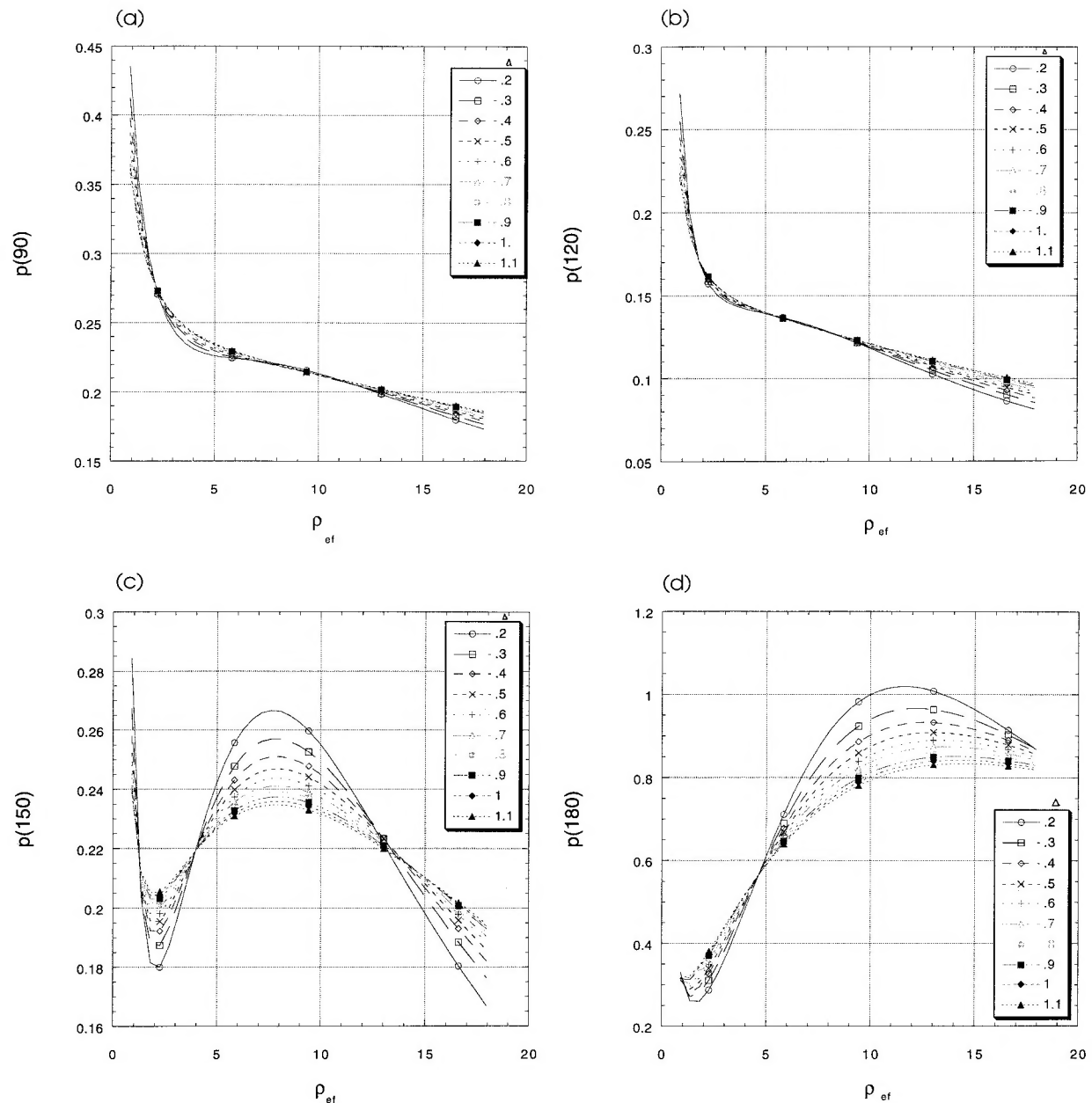


FIG. 2. Dependence of the phase function on the value of the effective size parameter $\rho_{ef} = 2\pi a_{ef}/\lambda$ at $m = 1.53 - 0.006i$, $\theta = 90^\circ$ (a), 120° (b), 150° (c), and 180° (d), and different values of the coefficient of variance of the particle size distribution $\Delta = 0.2(0.1)1.1$.

About 150 phase functions formed the background of the statistical analysis in this case. The results of computing the coefficient of variation of the phase function are represented in Fig. 3a. One can see that the CV has a minimum around a scattering angle of $\theta = 157^\circ$ and grows from 8% to 16% while the scattering angle changes from 90° to 180° . The value of the CV at the minimum is about 6%. Thus, the variability of the phase function in this case is smaller than that of the phase function related to the effective size parameter change (see Fig. 1a).

The coefficient of variation of phase functions of submicrometer aerosols with different values of the imaginary part of the refractive index $k = 0.001(0.0001)0.01$ at $\lambda = 0.7 \mu\text{m}$, $n = 1.53$, $a_{ef} = 0.2 \mu\text{m}$, and $\Delta = 0.7$ is represented in Fig. 3b. It increases with the scattering angle, but it is less than 5% at all scattering angles. This variation is small for most atmospheric optics applications and can be neglected.

It is possible to arrange the microphysical parameters of the submicrometer aerosol according to their influence on the phase functions at large scattering angles

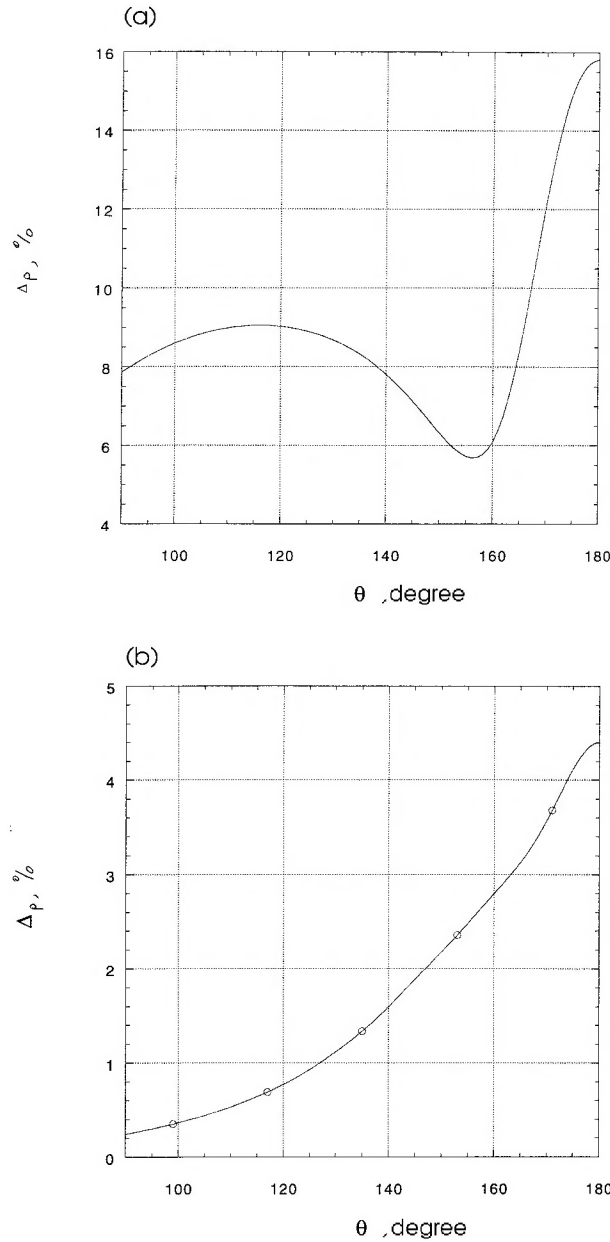


FIG. 3. (a) Dependence of the coefficient of variance of the phase function $\Delta_p = s_p(\theta)/\langle p(\theta) \rangle$ [see Eqs. (2) and (3)] on the scattering angle θ at $\lambda = 0.7 \mu\text{m}$, $\Delta = 0.7$, $a_{ef} = 0.2 \mu\text{m}$, $k = 0.006$, and different values of the real part of the refractive index $n = 1.45\text{--}1.6$. (b) Dependence of the coefficient of variance of the phase function Δ_p on the scattering angle θ at $\lambda = 0.7 \mu\text{m}$, $\Delta = 0.7$, $a_{ef} = 0.2 \mu\text{m}$, $n = 1.53$, and different values of the imaginary part of the refractive index $k = 0.001\text{--}0.01$.

in the following order: the effective radius of particles, the real part of the refractive index, the coefficient of variance of the particle size distribution, and the imaginary part of the refractive index.

We thus conclude that, despite the very high variability of atmospheric aerosol, the value of the phase function around the scattering angle of 150 degrees is

0.22 with an accuracy from 20% to 30% for the background atmospheric aerosol. This fact can be used in different applications, including atmospheric correction problems and aerosol optical thickness retrieval from space.

3. Error in determining aerosol optical thickness from space

The obtained results can be applied to estimating the error in determining aerosol optical thickness over the ocean from space. In the single scattering approximation (without including multiple light scattering and surface reflection effects), the reflection function $R(\mu, \mu_0, \psi)$ of the atmosphere over ocean is (Stephens 1994)

$$R(\mu, \mu_0, \psi) = \frac{\omega p_a(\theta)}{4(\mu + \mu_0)} \left\{ 1 - \exp \left[-\tau \left(\frac{1}{\mu} + \frac{1}{\mu_0} \right) \right] \right\}, \quad (6)$$

where ω is the single scattering albedo, $p_a(\theta)$ is the atmospheric phase function, $\mu = \cos \vartheta$, $\mu_0 = \cos \vartheta_0$, ϑ_0 is the solar angle, ϑ is the observation angle,

$$\cos \theta = -\mu\mu_0 + \sqrt{(1 - \mu^2)(1 - \mu_0^2)} \cos \psi$$

ψ is the azimuth angle, and τ is the optical thickness. From Eq. (6), it follows:

$$\tau = \frac{\mu_0\mu}{\mu_0 + \mu} \ln \left[1 - \frac{4(\mu + \mu_0)}{\omega p_a(\theta)} R(\mu, \mu_0, \psi) \right]^{-1}. \quad (7)$$

Equation (7) can be used to investigate the error in determining the optical thickness related to the uncertainties in the values of the single scattering albedo and the phase function (at least, in a first coarse approximation). It follows from Eq. (7):

$$\frac{\Delta \tau}{\tau} = K \sqrt{\frac{(\Delta \omega)^2}{\omega^2} + \frac{(\Delta p_a)^2}{p_a^2}}, \quad (8)$$

where

$$K = \frac{x}{(x - 1) \ln(1 - x)},$$

$$x = \frac{4(\mu + \mu_0)}{\omega p_a(\theta)} R(\mu, \mu_0, \psi). \quad (9)$$

It is well known (Smirnov et al. 1995; Gordon 1997) that the absorption of light by oceanic aerosols is extremely low ($\omega \approx 1$) in most cases. It follows that $(\Delta \omega)^2/\omega^2 \ll (\Delta p_a)^2/p_a^2$ and $\Delta \tau/\tau = K \Delta p_a/p_a$. One can see that the accuracy of the optical thickness determination over ocean depends mostly on the uncertainty in the value of the atmospheric phase function.

It is interesting that the coefficient K depends only on the parameter $x = 1 - \exp[-\tau(1/\mu + 1/\mu_0)]$ [see Eqs. (6) and (9)]. For atmospheric aerosols over the ocean, τ is less than 0.3 at $\lambda = 0.7 \mu\text{m}$ in most of cases

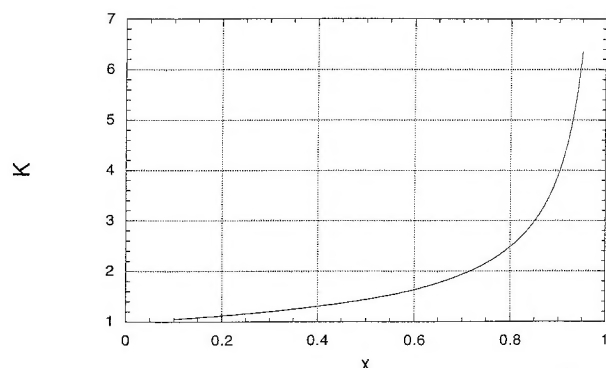


FIG. 4. Dependence of the amplification factor K on x according to Eq. (9).

(Smirnov et al. 1995). The value of $M = 1/\mu + 1/\mu_0$ changes from 2 to 4 for solar and observation angles less than 60° . This leads to the values of $x < 0.7$ and $K < 2$ (see Fig. 4). One can see that the error of the optical thickness determination can be two times larger than the uncertainty in the value of the atmospheric phase function.

Let us now consider the case of nadir measurements and a scattering angle of 150° . This angle will occur at the sun angle of $\vartheta_0 = 30^\circ$. The values of x and K in this case are $x \leq 0.5$ (at $\tau \leq 0.3$), $K \leq 1.5$ (see Fig. 4).

The atmospheric phase function can be presented in the following form (Liou 1992):

$$p_a(\theta) = \frac{p(\theta) + \xi p_m(\theta)}{1 + \xi}, \quad (10)$$

where $p(\theta)$ is the aerosol phase function, $p_m(\theta) = 0.75(1 + \cos^2\theta)$ is the molecular scattering function, and ξ is the ratio of the molecular optical thickness to the aerosol scattering optical depth.

From Eq. (10) it follows:

$$\frac{dp_a(\theta)}{p_a(\theta)} = C \frac{dp(\theta)}{p(\theta)}, \quad C = \frac{1}{1 + \xi \frac{p_m(\theta)}{p(\theta)}}. \quad (11)$$

Note that ξ decreases with the wavelength and is not smaller than 0.1 at $\lambda = 0.7$ for most cases (Bucholtz 1995). Here, C thus becomes less than 0.6 at $\theta = 150^\circ$, and the error of the optical thickness determination is about equal to the uncertainty in the aerosol phase function (20%–30% at a scattering angle of 150°). Therefore, the error of the optical thickness determination using Eq. (7) can be as large as 20%–30% within the framework of the considered algorithm.

It is interesting that the more precise analysis of Koepke and Quenzel (1979), which considered multiple light scattering and light reflection from ocean, gives about the same estimate for the value of $\Delta\tau/\tau$ (25%). However, only four Sahara dust aerosol size distributions were considered by Koepke and Quenzel (1979),

and another region of favorable angles was obtained (e.g., their favorable sun angles change from 50° to 65° at nadir measurements, not around 30° as in this paper, based on the simple single light-scattering approach). The author has no an explanation for these discrepancies.

4. Conclusions

The variability of the phase function of a submicrometer aerosol was studied in detail. It was found that the phase function of the atmospheric aerosol is the most stable around a scattering angle of 150° . This fact was used to estimate the error of determining the optical thickness determination over the ocean from space at this scattering angle within the framework of a single scattering approximation without accounting for the reflection of light from the ocean surface.

This error is about equal to the uncertainty in the value of the aerosol phase function. As already presented, the coefficient of variance of the phase function is minimal at scattering angles around 150° . This scattering angle is therefore favorable for remote sensing of turbidity from space, at least for small τ (for nadir measurements, the solar angle should be around 30°).

The paper of Koepke and Quenzel (1979) reported that the favorable sun angles should be in the region 50° – 65° for nadir measurements for four types of the Sahara dust aerosols (considering the surface reflection and multiple light scattering). In this case, the scattering angles are from 115° to 130° , and the coefficient of variance of the aerosol phase function is approximately two times larger than that at a scattering angle of 150° (see Fig. 1a).

The author calculated phase functions of four types of aerosols presented by Koepke and Quenzel (1979). All of them have a cross point at 147° . This is in accordance with Fig. 1 of this paper, Fig. 4 of Kaufman et al. (1994), Fig. 2 of Gordon and Zhang (1996), and Fig. 9 of Gordon (1997). Thus, in the single scattering approximation (very thin aerosol layers), the favorable scattering angles are around 150° .

It should be pointed out that this manuscript considers only variations along the coordinate axes of the four-dimensional parameter space (a_{ef} , Δ , n , χ). More complete coverage of the variable space is a subject for future work.

Acknowledgments. The author wishes to express his thanks to Prof. T. Nakajima, Dr. T. Takashima, and anonymous reviewers for valuable comments. He is deeply grateful to the staff members of NASDA's Earth Observation Research Center (Tokyo) for their encouragement throughout his work. Special thanks are due to Dr. E. P. Zege of the Institute of Physics in Minsk, who suggested this work.

This contribution was supported by the Science and

Technology Agency of Japan and NASDA's Earth Observation Research Center.

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